

Serial No. 10/530,554
Amendment dated April 30, 2007
Official Action dated November 30, 2006

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REMARKS

The Office Action dated November 30, 2006 has been carefully considered. Accordingly, the changes presented herewith, taken with the following remarks, are believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present amendment, claim 1 has been amended to clarify that ring-opening polymerization is conducted by bulk ring-opening polymerization in accordance with the teachings of the specification, for example, at paragraph [0053] and in the Examples. Claim 1 has also been amended to more clearly recite that the overall proton concentration in the cyclic ester is calculated out on the basis of the total amount of hydroxycarboxylic compounds as impurities in the cyclic ester, water contained as impurities in the cyclic ester and water added to the cyclic ester. Support for these limitations may be found in the specification at paragraphs [0074] and [0095]. Claim 2 has been canceled from the application, Claim 3 has been amended to change its dependency from canceled claim 2 to claim 1, and Claims 14 and 16 have been amended for matters of form. Since these changes do not involve any introduction of new matter, entry of the amendments is believed to be in order and is respectfully requested.

In the Official Action, claims 1-6 and 13-20 were rejected under 35 U.S.C. §103 (a) as being unpatentable over Shinoda et al (US 5,412,067) in view of Howelton et al (US 5,342,918). The Examiner asserted that Shinoda et al disclose a preparation process for a polyester with a desirable molecular weight from cyclic esters or their mixtures, wherein impurities such as water and hydroxy carboxylic acids are controlled to a total amount of less than 100 ppm to produce a polyester with a desirable molecular weight. The Examiner relied on Howelton et al as teaching the addition of water in a ring-opening polymerization. The Examiner concluded it would have been obvious to use the purified cyclic ester in order to

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obtain polyester with a desirable molecular weight according to the teachings of Shinoda et al and to add water to the polymerization reaction system to start and/or increase the rate of polymerization as shown by Howelton.

However, as will be set forth in detail below, Applicants submit that the production processes defined by claims 1, 3-6 and 13-20 are not rendered obvious over the combined teachings of Shinoda et al and Howelton et al. Accordingly, the rejection is traversed and reconsideration is respectfully requested.

More particularly, as defined by claim 1, the present invention relates to a process for producing an aliphatic polyester by subjecting a cyclic ester to bulk ring-opening polymerization. The process comprises adding water to a cyclic ester purified to the extent that a water content is at most 60 ppm to control an overall proton concentration in the cyclic ester, thereby controlling at least one physical property among melt viscosity, molecular weight and yellowness index of the resulting aliphatic polyester. The overall proton concentration in the cyclic ester is calculated out on the basis of the total amount of hydroxycarboxylic compounds as impurities in the cyclic ester, water contained as impurities in the cyclic ester and water added to the cyclic ester.

Shinoda et al disclose a polyester preparation process which includes adding a hydroxyl compound as a molecular weight regulator to a reaction system and conducting ring-opening polymerization of a cyclic ester compound. Shinoda et al teach fixing an amount of the hydroxyl compound to be added to the reaction system on the basis of the amount of free carboxylic acid contained in the cyclic ester compound (claim 1). The hydroxyl compound is one or more compounds selected from alcohol, hydroxycarboxylic acid and saccharide (claim 2). The alcohol is one or more monohydric straight chain saturated aliphatic alcohols having 12-18 carbon atoms (claim 3). In the Examples of

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Shinoda et al, lauryl alcohol and lactic acid are the hydroxyl compounds added to the reaction systems.

However, the process for producing an aliphatic polyester according to the present invention is significantly different from the polyester preparation process of according to Shinoda et al. The basis of the invention according to Shinoda et al is to previously estimate the amount of free carboxylic acid contained as impurities in the cyclic ester compound raw material, and to fix the amount of the hydroxyl compound to be added to the reaction system on the basis of the measured value. By carrying out ring-opening polymerization of the cyclic ester compound according to the preparation process of Shinoda et al, polyester having a molecular weight which is accurately controlled within a desired range can be consistently prepared even though the amount of the free carboxylic acid contained in the cyclic ester compound raw material is varied for each batch.

On the other hand, the characteristic feature of the present invention resides in that water is added to a purified cyclic ester to control an overall proton concentration in the cyclic ester. The overall proton concentration in the cyclic ester is calculated out on the basis of the total amount of hydroxycarboxylic compounds as impurities in the cyclic ester, water contained as impurities in the cyclic ester and water added to the cyclic ester. The following differences demonstrate the distinctions between the process according to the present invention as compared with the process according to Shinoda et al.

First, Shinoda et al do not teach, suggest or recognize that there is a close correlation between the overall proton concentration in the cyclic ester and the molecular weight and melt viscosity of the polyester obtained by conducting ring-opening polymerization of the cyclic ester. Second, in the process of Shinoda et al, the hydroxyl compound (for example, lauryl alcohol) is added to the reaction system, and no water is added. To the contrary, Shinoda et al describe that the cyclic ester compound is preferably dehydrated as much as

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possible before subjecting the compound to the polymerization reaction, and in order to accurately control the molecular weight of polyester having a molecular weight of 100,000 or more in particular, the moisture content of the cyclic ester compound is preferably 100 ppm by weight or less (column 6, lines 39-50). As Shinoda et al teach that the amount of water contained in the cyclic ester compound is reduced as much as possible in order to accurately control the molecular weight of the resulting polyester, quite naturally, in the process of Shinoda et al, the addition and mixing of water are positively excluded in the ring-opening polymerization of the cyclic ester compound.

On the other hand, in the process according to the present invention, water is added to the cyclic ester having a water content of at most 60 ppm to change the overall proton concentration in the cyclic ester. The overall proton concentration is a value calculated out on the basis of the total amount of hydroxycarboxylic compounds contained in the cyclic ester and water. When the overall proton concentration in a purified cyclic ester is controlled by adding water to the cyclic ester according to the invention, the physical properties of the formed polymer, such as melt viscosity, molecular weight and yellowness index, can be precisely controlled. It has been surprisingly shown that when water is used as a molecular weight control agent, the reaction efficiency of the ring-opening polymerization is increased and the content of volatile matter comprising a remaining monomer as a main component is reduced. In other words, when water is used as the molecular weight control agent according to the present invention, a polymer having a high molecular weight and a high melt viscosity can be synthesized while reducing the amount of the volatile matter (residual monomer) to a low level.

Thus, Applicants have determined a close correlation between the overall proton concentration in the cyclic ester and the melt viscosity and/or molecular weight of the polymer and the presently claimed processes advantageously employ such. More

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specifically, such a correlation as described can be clarified by conducting ring-opening polymerization under the same polymerization conditions (for example, reaction vessel, polymerization temperature, polymerization time, the kind and purification degree of the monomer used, etc.) except that the overall proton concentration in the cyclic ester is changed by changing the amount of water added, measuring the melt viscosities, molecular weights and yellowness indexes of the resulting aliphatic polyesters and carrying out regression analysis using the measured results as a database. For example, the overall proton concentration was changed by adding water to glycolide, and the melt viscosity, weight average molecular weight and yellowness index of polyglycolic acid obtained by ring-opening polymerization of the glycolide were measured. As a result, applicants have demonstrated that these respective physical properties are related to the overall proton concentration.

Applicants particularly direct the Examiner's attention to the resulting data from such processes which are summarized in Table 1 of the present specification. For example, when 1-dodecyl alcohol (lauryl alcohol) is added as the molecular weight control agent to conduct ring-opening polymerization of glycolide as taught by Shinoda et al and as shown in Comparative Example 3 in Table 1 at page 43 of the present specification, the weight average molecular weight of the resulting polyglycolic acid can be controlled with a measure of accuracy. However, the amount of volatile matter (residual monomer) contained in the polyglycolic acid is relatively high, i.e., 0.55% by weight. Since the polyglycolic acid in Comparative Example 3 contains such a high amount of volatile matter, the melt viscosity thereof is markedly low (1,560 Pa·s) though it is a high-molecular weight ($M_w = 181,000$) polymer.

In contrast, when glycolide, to which water is added as the molecular weight control agent so as to control the overall proton concentration to a predetermined value according to

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the present invention, is used to conduct ring-opening polymerization as shown in Examples 1-4 in Table 1 at page 43, polyglycolic acid having desired melt viscosity and weight average molecular weight can be obtained. In addition, the polyglycolic acid has an extremely low amount of volatile matter.

Melt viscosity is a physical property used to preset molding or forming conditions of the aliphatic polyester and also used for predicting the mechanical strength and the like of the resulting molded or formed product. Accordingly, from the viewpoint of production techniques, it is important and advantageous to produce an aliphatic polyester having a targeted melt viscosity. The molecular weight of the aliphatic polyester is also a physical property used to preset molding or forming conditions thereof and contributes to the mechanical strength of the resulting molded or formed product. The yellowness index of the aliphatic polyester is an index indicating the quality of the aliphatic polyester and is an important physical property for obtaining a product of a desired color.

When ring-opening polymerization is conducted without adding water to the purified cyclic ester, unreacted monomer tends to remain in the formed polymer (See Comparative Example 1 in Table 1 at page 43). As the content of volatile matter comprising the remaining monomer as a main component increases, the melt viscosity of the resulting polymer is reduced. In addition to deterioration of polymer quality, the yellowness index of the polymer also increases. Thus, it is difficult to control the melt viscosity or the like of the resulting polymer by mere control of the degree of purification of the cyclic ester.

When a higher alcohol is used as a molecular weight control agent, as taught by Shinoda et al, the amount of the volatile matter can be somewhat reduced. However, the reduction is not sufficient, and a polymer containing the remaining monomer in a considerable amount is formed (Comparative Example 3 in Table 1). Therefore, it is difficult to precisely control the melt viscosity of the formed polymer by using the higher alcohol.

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More specifically, it is possible to control the molecular weight (for example, weight average molecular weight) of the resulting aliphatic polyester using the higher alcohol, but since the polymer contains the remaining monomer in a relatively high amount, the melt viscosity thereof may be low in some cases. In this case, the melt viscosity of the polymer is affected by not only the molecular weight thereof, but also the amount of the remaining monomer. Further, when a higher alcohol is used as the molecular weight control agent, it is difficult to lessen the yellowness index of the resulting polymer. On the other hand, when the overall proton concentration in the cyclic ester is changed by addition of water, as in the processes of the present invention, the melt viscosity and molecular weight of the polymer formed can be controlled within the desired respective ranges while reducing the amount of the volatile matter (residual monomer) to a low level (Examples 1-4 in Table 1).

Disadvantages of the use of a higher alcohol as a molecular weight control agent as taught by Shinoda et al are described above. In addition, a higher alcohol such as lauryl alcohol is expensive and is a viscous material which tends to remain in a charging device when charged into a polymerization reaction system, resulting in material loss. In addition, when higher alcohol is used in the production of an aliphatic polyester on an industrial scale increased production costs are required in that (1) greater amounts are used, (2) a tank and a weighing device, which are temporarily used for charging the alcohol into a polymerization reactor, are enlarged in size, (3) the apparatus must be provided with safety measures because the alcohol is combustible, (4) cleaning of the apparatus is required after the apparatus are used, and (5) cleaning liquids used in the cleaning of the apparatus must be treated. Further, when a higher alcohol is added to a cyclic ester, the higher alcohol is introduced into the aliphatic polyester polymer structure because the higher alcohol also acts as an initiator, and the physical properties of the resulting aliphatic polyester are changed. Furthermore, the higher alcohol has insufficient solubility in the cyclic ester, and consequently the ring-

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opening polymerization reaction takes place unevenly, thereby preventing precise control of the molecular weight and melt viscosity of the resulting polymer. These disadvantages are also encountered when another hydroxyl compound is used in place of a higher alcohol.

Shinoda et al neither teach nor suggest addition of water to control the overall proton concentration in the cyclic ester, thereby controlling the melt viscosity and molecular weight of the resulting aliphatic polyester. Water is not only inexpensive but also allows simplification and scale-down of the production apparatus. When water is used as a molecular weight control agent, it is unnecessary to conduct cleaning of the apparatus, to treat a cleaning liquid, and the like. Accordingly, the presently claimed process making use of water is advantageous with respect to energy saving, cleaning and environmental protection.

The deficiencies of Shinoda et al are not resolved by Howelton et al. That is, Howelton et al relate to a ring-opening reaction of cyclic lactam such as caprolactam and laurolactam, and provide no teaching or suggestion relating to a ring-opening reaction of a cyclic ester such as glycolide and lactide. More particularly, Howelton et al disclose carboxyl-terminated polyetheramides made by capping polyetheramides (aminoacids) with cyclic lactams (column 3, lines 25-28). Cyclic lactams have the structure of the formula shown on column 5, and are not cyclic esters.

EXAMPLES 1-4 of Howelton et al describe that

"The reactor was charged with laurolactam and various carboxylic acids/amino acids (as specified in Table 1), 50 ml of deionized water and 0.25 grams of IRGANOX 1098. The reactants and water were charged then purged with nitrogen for 15 minutes followed by heating at 250 °C. for 2 hours to allow ring opening of the lactam with water. Water was slowly vented off and reactants were held at 250 °C. for one additional hour." (Column 6, lines 39-45)

As described above, in Howelton et al, water is added to the reaction system to allow ring opening of the cyclic lactam. Howelton et al neither disclose nor suggest that water may

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be added to a cyclic ester as presently claimed or to control the molecular weight, melt viscosity or yellowing of a ring-opening polymer of the cyclic lactam.

On the other hand, in the process according to the present invention, the ring-opening polymerization of the cyclic ester is conducted by using a ring-opening polymerization catalyst in accordance with a method known *per se* in the art. Accordingly, one of ordinary skill in the art would have no reason to employ water addition along the lines of Howelton et al in the process of Shinoda et al. On the other hand, in the process according to the present invention, water is added to control the overall proton concentration in the cyclic ester to obtain a desired combination of physical properties and Howelton et al provide no teaching or suggestion in this regard.

In view of the foregoing, Applicants submit that the combination of Shinoda et al and Howelton et al does not lead to the claimed invention. In order for a combination of references to render an invention obvious, the combination of the teachings of all or any of the references must suggest, expressly or by implication, the possibility of achieving further improvement by combining such teachings along the line of the invention. *In re Sernaker*, 702 F.2d 989, 217 USPQ 1 (Fed. Cir. 1983). Applicants find no suggestion, express or implied, in the combination of Shinoda et al and Howelton et al relating to the possibility of achieving further improvement by combining such teachings along the lines of the present invention. Shinoda et al and Howelton et al neither teach nor suggest that physical properties such as a melt viscosity and a molecular weight of an aliphatic polyester can be precisely controlled by adding water as presently claimed.

It is therefore submitted that the processes for producing an aliphatic polyester defined by claims 1, 3-6 and 13-20 are nonobvious over and patentably distinguishable from the cited combination of references, whereby the rejection has been overcome. Reconsideration is respectfully requested.

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Claims 7-9 were rejected under 35 U.S.C. 103 (a) as being unpatentable over Shinoda et al in view of Howelton et al as applied to claims 1-6 and 13-20 above, and further in view of Early et al (US 6,437,565). The Examiner relied on Early et al as disclosing the use of regression analysis in order to determine physical properties of a composition. The Examiner asserted that it would have been obvious to use regression analysis as taught by Early et al in order to find an optimum amount of proton concentration for obtaining polyester with desirable properties.

This rejection is traversed. The deficiencies of Shinoda et al and Howelton et al are discussed above. Early et al fail to resolve such deficiencies. Further, Early et al disclose a method for determining at least one physical property of a material, for example crosslink density of a rubber, by NMR measurement by relating a ratio of a spin lattice relaxation time to a spin-spin relaxation time to known values of the physical property. However, Applicants find no teaching by Early et al relating to a control step as recited in any of claims 7-9 wherein the overall proton concentration in a cyclic ester is controlled on the basis of a regression expression between a predetermined overall proton concentration in the cyclic ester and a physical property value. Further, Applicants find no teaching or suggestion by Early et al, of Shinoda et al or Howelton et al which would suggest to one of ordinary skill in the art that proton concentration is a result effective variable and can be controlled by addition of water to provide desired properties. Accordingly, these references do not combine to render claims 7 - 9 obvious.

It is therefore submitted that the processes for producing an aliphatic polyester defined by claims 7-9 are nonobvious over and patentably distinguishable from the cited combination of references, whereby the rejection has been overcome. Reconsideration is respectfully requested.

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Finally, claims 10-12 were rejected as being obvious and unpatentable over Shinoda et al in view of Howelton et al and further in view of the *Handbook of Thermoplastic Polymers*, Chapter 2, at pages 80-94 (HTP). The Examiner asserted HTP discloses a ring-opening polymerization process can be conducted in a closed volume because the process does not require evacuation of byproducts, referring to pages 90-94 of HTP. The Examiner concluded it would have been obvious to conduct the polymerization process disclosed by Shinoda et al in view of Howelton et al inside closed tubes as a simple polymerization reactor. Regarding claim 12, the Examiner asserted that solid-state polymerization is well known, referring to pages 80-82 of HTP.

This rejection is traversed. Applicants find no teaching or suggestion by HTP for resolving the deficiencies of Shinoda et al and Howelton et al as discussed above. To the contrary, HTP indicates at page 94 that extreme care should be taken to insure that the cyclic dimer will contain neither water nor linear oligomers. Accordingly, the water addition step required by the present claims is opposite to the teachings of HTP. Thus, HTP provides no teaching or suggestion of the sequence of process steps recited in claim 10 including the step of adding water to the purified cyclic ester. Further, the solid state polymerization referred to by the Examiner at pages 80-82 of HTP is directed to production of poly(ethylene terephthalate), and not a ring-opening polymerization as required by the present claims. Thus, the cited combination does not render claims 10 - 12 obvious.

It is therefore submitted that the processes for producing an aliphatic polyester defined by claims 10-12 are nonobvious over and patentably distinguishable from the cited combination of references, whereby the rejection has been overcome. Reconsideration is respectfully requested.

Finally, with respect to German Reference 131,064 cited in the Information Disclosure Statement filed August 2, 2006, Applicants note that this reference discloses a

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process for conducting solution polymerization of glycolide and analogues thereof at a temperature of 50°C or lower, in which an alcohol solution of an alkali metal hydroxide and/or an alkali metal or alkaline earth metal alcoholate is used as a strongly basic catalyst, and a polar solvent miscible with water is used as a solvent (claim). Example 1 of German Reference 131,064 describes that glycolide was dissolved in acetone and stirred with alcoholic KOH while keeping a temperature of 20°C, and that a polymer formed was washed with water and methanol and dried. Finally, German Reference 131,064 describes that glycolide having a high purity is used (page 2, lines 22-26).

It is believed that the above represents a complete response to the Official Action and places the present application in condition for allowance. Reconsideration and an early allowance are requested.

Respectfully submitted,



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